Scalapino, Bull. Am. Phys. Soc. 13, 475 (1968).

[4]A. T. Larkin and Yu. N. Ovchinnikov, Zh. Eksperim. i Teor. Fiz. 53, 2159 (1967) [translation: Soviet Phys. -JETP 26, 1219 (1968)].

[5]A small magnetic field can be applied to remove the pair-transfer part of the dc current so that I_{qp} can be directly measured.

[6]D. Rogovin, thesis, University of Pennsylvania, 1969 (unpublished).

[7]J. L. Stewart, Proc. IRE 14, 1539 (1954). The spectrum of fluctuations is flat out to frequencies of order $(CR_D)^{-1}$, where C is the junction capacitance. Since $\Delta\omega \ll (CR_D)^{-1}$, the fraction of the total voltage fluctuation contributing to the linewidth is much less than unity so that the resulting line is essentially Lorentzian, as in the analogous case of motional or exchange narrowing in magnetic resonance.

[8]B. D. Josephson, Advan. Phys. 14, 419 (1965). D. Rogovin has carried out a direct calculation of $\langle I(t)I(0)\rangle$ which shows that at finite temperature, the term $\sigma_1'(V_0)V_0\cos\varphi$ enters the expression for the pair-fluctuation part of the current power spectrum (Ref. 6).

[9]In the zero-temperature limit $\Delta\omega$ approaches $(8e^3/\hbar^2)R_D{}^2I(V_0)$, which is a factor of 2 smaller than the result obtained in Ref. 1. In that paper, an additional contribution to the fluctuations arises from the reservoirs which were formally introduced to simulate the leads to the junction. We believe that the correct representation of the external coupling does not contain these fluctuations. M. J. Stephen concurs (private communication).

[10]D. N. Langenberg, D. J. Scalapino, and B. N. Taylor [Proc. IEEE 54, 560 (1966)] have discussed self-induced steps and other properties of radiating Josephson junctions.

[11]N. R. Werthamer and S. Shapiro, Phys. Rev. 164, 523 (1967).

SUPPRESSION OF THE KONDO MANY-BODY SCATTERING EFFECT

D. Gainon and A. J. Heeger*
Institut de Physique de la Matière Condensée, University of Geneva, Geneva, Switzerland
(Received 5 May 1969)

An experimental study of the electrical resistivity in dilute Cu:Mn doped with Pt impurities is presented. The results indicate a suppression of the Kondo divergence in the conduction-electron scattering amplitude as a result of the spin-orbit interaction at the Pt impurities. The results are interpreted in terms of a reduction in the spin "memory" time.

In 1964, Kondo[1] demonstrated the existence of a logarithmic divergence in perturbation theory for the scattering of conduction electrons from magnetic impurities in metals. Since that time, there has been considerable research directed toward the goal of understanding the physical origin of the many-body scattering as well as toward a determination of the ground state of this coupled many-body system. Although we still do not have a satisfactory theory for temperatures less than the divergence temperature[2]

$$T_K = (D/k)\exp[-1/|J|\rho], \qquad (1)$$

existing experimental and theoretical work[3] indicates that the ground state is a many-body singlet in which nonperturbative spin correlations are built up in the electron gas in the vicinity of the impurity. These spin correlations may be viewed as fundamentally arising from an indirect electron-electron interaction[4] via the magnetic impurity. A first electron scatters from the impurity leaving the spin flipped so that subsequent scattering of a second electron depends on the fact that the first event has already taken place. From this point of view, the question of the "memory" of the impurity spin in the intermediate state takes on special significance. If this memory is made sufficiently short either perhaps by intrinsic coupling of the impurity to the conduction electrons (i.e., spin fluctuation effects[5]) or by direct relaxation of the impurity spin to the lattice, one may expect a suppression of the indirect interaction and the associated logarithmic divergence with the result that the many-body spin correlations will not form. In this Letter we present electrical-resistivity data which yield experimental evidence of such suppression of the Kondo effect in the system Cu:Mn due to spin-orbit scattering from (heavy) Pt impurities.

The temperature dependence of the resistivity for Cu:Mn (110 ppm of Mn) doped with 0, 0.1, 0.3, 0.5, 1, 2, and 5 at.% Pt has been studied between 1.5 and 4.2°K. Figure 1 shows the results for the lowest concentrations. Platinum was chosen because of its relatively good solubility in Cu as well as its high Z and thus large value of

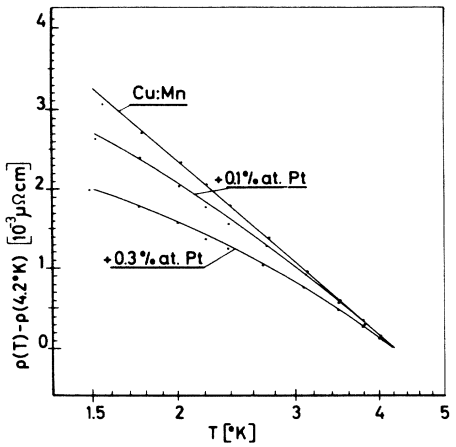

FIG. 1. The temperature-dependent contribution to the low-temperature resistivity in dilute (110 ppm Mn) Cu:Mn doped with Pt impurities. The resistivity at 4.2°K is subtracted to remove all temperature-independent contributions. The solid curves represent Eq. (2) of the text.

the spin-orbit coupling constant λ. For the undoped Cu:Mn one sees the expected characteristic logarithmic temperature dependence of $\rho(T)$. However, addition of as little as 0.1% Pt causes a marked deviation from the logarithmic dependence in such a way as to significantly decrease the anomalous scattering. The data in Fig. 1 are $\rho-\rho_{4.2}$ plotted as a function of T where $\rho_{4.2}$ is the value of the resistivity at 4.2°K. This subtraction is made to remove the temperature-independent contribution due to the Pt and thereby show explicitly the temperature dependence of the anomalous scattering from the magnetic impurities. The resistivity of a sample of Cu:Pt (Pt concentration of 1 at.%) was separately checked and found to be temperature independent below 4.2°K in agreement with expectations. The suppression of the Kondo effect is clearly shown in the data. It is impossible to explain the reduction in the magnitude and the induced curvature of $\Delta\rho$ in terms of a change in the Kondo temperature. For Cu:Mn, T_K has been shown to be of order 0.05°K.[6] Consequently, to explain the present results would require an increase in T_K by two orders of magnitude with addition of a fraction of a percent of Pt. Even such an unlikely assumption leads to difficulty for the maximum slope of $\rho(T)$ should occur at $T \simeq T_K$ with a saturation at still lower temperatures. Moreover, the sensitivity of the results to the spin-orbit coupling constant rather than the valence difference (see below) is inconsistent with a change in T_K.

The samples were made by arc melting, using high-purity starting material. To insure constant Mn concentration all samples were made from a 0.2-at.% Cu:Mn master alloy. The samples to be measured were subsequently drawn into 0.2-mm-diam wire (one to three meters in length), coiled, and placed directly into liquid helium. The resistivity was measured using the standard four-terminal technique with a constant current of about 0.7 mA. The relative accuracy for the temperature dependence of a single sample is better than +0.05% as a result of the long wires and relatively large resistance. The absolute accuracy is estimated to be ±5% with the dominant source of error being the determination of the wire diameter.

The solid curves in Fig. 1 are a fit of the expression

$$\rho(T) = A \ln[T^2 + \theta^2]^{1/2} \qquad (2)$$

to the data. The agreement of Eq. (2) with the experimental results is in all cases within the small scatter of the individual points. Other expressions [e.g., $\ln(T+\theta)$] were found not to provide as good a description of the data. The curves in Fig. 1 assume a fixed value of the coefficient A (i.e., of the s-d exchange constant J) equal to that in the undoped Cu:Mn. The different curves are thus obtained by choosing the single parameter θ for best agreement with the experimental results. The resulting dependence of θ upon the Pt concentration is shown in Fig. 2. One sees an initial very steep rise ($d\theta/dc \cong 11$°K/at.%), an apparent saturation, and a much slower subsequent increase.

In order to explicitly demonstrate the importance of the spin-orbit interaction a sample of Cu:Mn doped with 1% Ge was studied. The temperature-dependent contribution to the resistivity

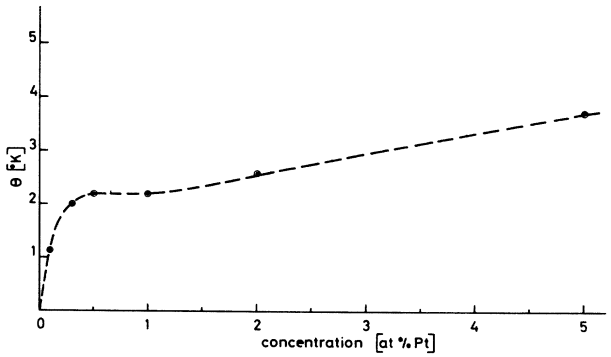

FIG. 2. The dependence of θ on the concentration of Pt. The values are obtained by adjusting θ [in Eq. (2) of the text] to obtain a best fit to the data of Fig. 1.

deviated by at most 10% from that of the undoped Cu:Mn. This is in agreement with earlier studies which showed essentially no suppression of the logarithmic term due to low-Z, and hence small-λ, impurities.[7] Since the residual resistivity of Ge in Cu (3.75 $\mu\Omega$ cm/at.%)[8] is larger than that of Pt in Cu (2.03 $\mu\Omega$ cm/at.%),[9] the negligible effect of the Ge impurities on the Kondo scattering provides good evidence that the spin-orbit interaction is the major source of the suppression.

An expression of the form of Eq. (2) was predicted by Suhl[10] for the situation when the Ruderman-Kittel-Kasuya-Yosida interaction between the magnetic impurities is important. Although Suhl's theory is not directly appropriate to the present results as evidenced by the good logarithmic dependence (i.e., $\theta \simeq 0$) of the undoped dilute Cu:Mn samples, the concept of a composite intermediate-state density which is a convolution of the density of the additional electron or hole with the density of all states having the impurity spin flipped may nevertheless be valid. However, in the present case the reservoir to which the spin is coupled is not the Ruderman-Kittel-Kasuya-Yosida system, but the conduction electrons themselves (via intrinsic spin fluctuation effects) or the lattice (via relaxation of the impurity spin). It has in fact been experimentally demonstrated[11] that because of the s-d admixture, the long tails on the Mn wave function can probe the spin-orbit interaction at distant impurities, thus leading to a finite relaxation rate in the electron spin resonance. This effect can be expected to shorten significantly the spin-memory time especially with addition of the high-Z Pt impurities. Alteration of the intrinsic spin-fluctuation memory time would seem to be a smaller effect since such spin-fluctuation phenomena result from the finite virtual level width via the impurity d-state density at the Fermi surface.[12] It is difficult to see how the spin-orbit interaction at the Pt sites can have a strong effect on the virtual-state density. However, detailed understanding must await a proper theoretical treatment of the effect of both spin fluctuations and relaxation on the Kondo many-body scattering.[13]

The existence of a finite θ in Eq. (2) implies a drastic reduction in the Kondo divergence temperature:

$$T_K^2 = T_{K0}^2 - \theta^2, \tag{3}$$

where T_{K0} is given by Eq. (1) assuming that J does not change. Clearly when $\theta > T_{K0}$, the divergence in perturbation theory is removed so that the nonperturbative spin correlations associated with the singlet ground state will never form.

Finally we would like to comment on the differences (and similarities) with respect to the effect of impurity scattering on the critical temperature T_c in superconductors. In superconductivity, as long as time-reversal invariance is maintained (i.e., the impurities are nonmagnetic), the effect of impurity scattering is to alter the detailed form of the conduction-electron wave functions. The indirect interaction via the phonons is not changed, and it is possible to construct from the time-reversed states a pairing theory of dirty superconductors[14] which predicts little change in T_c. In the present case, where the indirect electron-electron interaction takes place via the impurity spin, the situation is quite different. Because of the s-d admixture the spin is in a strict sense a part of the electronic system. As a result, the spin-orbit interaction at the Pt sites has an effect on the indirect interaction itself with a correspondingly large reduction in T_K.

Discussions of the effect of relaxation on the Kondo scattering with Dr. B. Giovannini, Dr. A. Zawadowski, and Professor H. Suhl are gratefully acknowledged. The Swiss National Science Foundation provided financial support for the experimental work.

---

*John Simon Guggenheim Fellow 1968-1969. Permanent address: Department of Physics, University of Pennsylvania, Philadelphia, Pa.

[1]J. Kondo, Progr. Theoret. Phys. (Kyoto) 32, 37 (1964).

[2]In Eq. (1) D is a cutoff representing the momentum dependence of J, J is the s-d exchange coupling constant, and ρ is the host metal density of states per atom.

[3]For references see M. D. Daybell and W. Steyert, Rev. Mod. Phys. 40, 380 (1968); A. J. Heeger, L. B. Welsh, M. A. Jensen, and G. Gladstone, Phys. Rev. 172, 302 (1968).

[4]A. J. Heeger (to be published).

[5]M. Levine and H. Suhl, Phys. Rev. 171, 567 (1968).

[6]I. A. Campbell, J. P. Compton, I. R. Williams, and G. V. H. Wilson, Phys. Rev. Letters 19, 1319 (1967).

[7]W. B. Muir and P. Elliston, in Proceedings of the Eleventh International Conference on Low Temperature Physics, St. Andrews, Scotland, 1968, edited by J. F. Allen, D. M. Finlayson, and D. M. McCall (St. Andrews University, St. Andrews, Scotland, 1968), p. 1254.

[8]F. J. Blatt, Phys. Rev. 108, 285 (1957).

[9]M. T. Beal-Monod, Phys. Rev. 164, 360 (1967).

[10]H. Suhl, Phys. Rev. Letters 20, 656 (1968).

[11]J. A. McElroy and A. J. Heeger, Phys. Rev. Letters 20, 1481 (1968).

[12]B. Giovannini and A. J. Heeger, to be published.

[13]The form of the curve in Fig. 2 suggests the presence of two mechanisms, the first having a large initial slope but saturating above 0.3-at.% Pt., and the second having an essentially linear behavior all the way to 5% Pt, but with a smaller slope. It is tempting to try to attribute these to the two mechanisms suggested in the text (i.e., relaxation and spin fluctuations), but further work is needed before a meaningful separation can be made.

[14]P. W. Anderson, J. Phys. Chem. Solids 11, 26 (1959).

BAND STRUCTURE AND ^{197}Au NUCLEAR-MAGNETIC-RESONANCE STUDIES IN $AuAl_2$, $AuGa_2$, AND $AuIn_2$ *

A. C. Switendick and Albert Narath
Sandia Laboratories, Albuquerque, New Mexico 87115
(Received 22 May 1969)

Measurements of the ^{197}Au Knight shift in $AuAl_2$, $AuGa_2$, and $AuIn_2$ are reported. In contrast to the strong temperature dependence of the ^{71}Ga Knight shift, only a weak temperature dependence of the ^{197}Au Knight shift in $AuGa_2$ is observed. Electronic energy-band calculations for these compounds yield differences which provide an improved model for interpreting these and other experimental results.

Considerable attention has recently been focused on the intermetallic compounds $AuAl_2$, $AuGa_2$, and $AuIn_2$.[1-4] This interest derives in part from the fact that samples with high residual-resistance ratios may be prepared, enabling Fermi-surface studies,[3-5] and in part from the unusual temperature dependence of the ^{71}Ga Knight shift in $AuGa_2$.[1] The lack of any temperature dependence of the ^{27}Al and ^{115}In Knight shifts in $AuAl_2$ and $AuIn_2$, respectively, coupled with electronic specific-heat[6] and de Haas–van Alphen (dHvA) Fermi-surface studies which "give no indication of differences which pointedly would distinguish any one from the other or, for that matter, from what would reasonably be expected from a free-electron-like model"[1] constitutes the $AuGa_2$ dilemma. Although there seems to be evidence that $AuGa_2$ is in some way different (e.g., Knight shift, thermoelectric power[7]) from the other two, there are properties (dHvA orbits, F_1 and F_2 in the notation of Ref. 5) for which $AuAl_2$ appears dissimilar from the other two.

In this note we report two new results for this series of compounds: (1) experimental results for the ^{197}Au Knight shifts for all three compounds, and (2) preliminary results of band-structure calculations which give a consistent interpretation of many of the observed similarities and dissimilarities and which offer a significant improvement over the free-electron model, which predicts no significant differences between any of the compounds. We offer these preliminary results in the hope that they may aid further interpretation and as a basis for additional refinement of the model.

The ^{197}Au resonances were detected in 300-mesh powdered samples by means of spin-echo techniques. The 60-kOe external field was provided by a NbZr superconducting solenoid. Adequate signal-to-noise ratios were achieved by averaging the echo intensities with a Fabri-tek 952/1062 high-speed digital signal averager. Knight shifts were calculated from the experimental frequency/field ratios using the reference ratio $\nu/H = 0.072\,923$ kHz/Oe. At 1.2°K the resulting shifts for $AuAl_2$ and $AuIn_2$ were $(+0.94 \pm 0.05)\%$ and $(+1.49 \pm 0.05)\%$, respectively. For $AuGa_2$ we obtained the following values:

$T = 1.2$°K, $K = (+1.54 \pm 0.02)\%$;

$T = 4.0$°K, $K = (+1.52 \pm 0.02)\%$;

$T = 27$°K, $K = (+1.53 \pm 0.03)\%$;

$T = 76$°K, $K = (+1.50 \pm 0.05)\%$;

$T = 232$°K, $K = (+1.34 \pm 0.10)\%$.

The absence of a strong temperature dependence for the ^{197}Au Knight shift is particularly noteworthy. In the same temperature range the ^{71}Ga shift is known to vary from -0.13% at 4°K to $+0.45\%$ at 230°K.[1] It follows that those states at the Fermi level which give rise to the positive ^{71}Ga Knight shift in the high-temperature region have little if any gold s character. The shifts in $AuGa_2$ and $AuIn_2$ have magnitudes which are similar to the shift in gold metal (1.65%).[8] In contrast, the $AuAl_2$ value is significantly smaller. We have also measured the ^{197}Au spin-lattice re-